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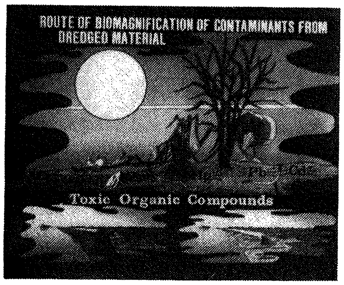

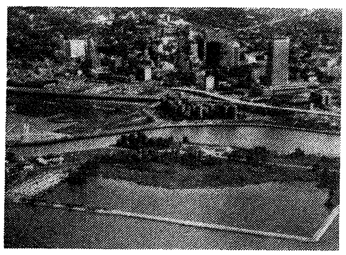

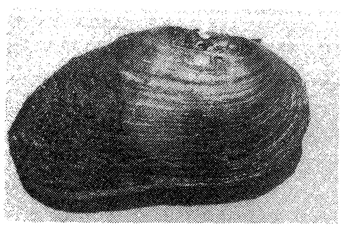

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MISCELLANEOUS PAPER EL-90-24

DISTRIBUTION OF PCB AND PESTICIDE CONTAMINANTS IN THE VICINITY OF THE TIMES BEACH CONFINED DISPOSAL FACILITY, BUFFALO, NEW YORK

by

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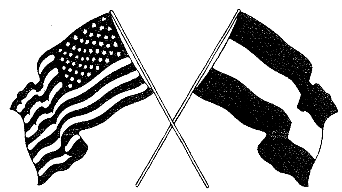

WES

December 1990

Final Report

Joint Research under the Auspices of
The United States/The Netherlands
Memorandum of Understanding Concerning
Dredging and Related Technology

Prepared for US Army Engineer District, Buffalo
Buffalo, New York 14207-3199

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY *(Leave blank)*

2. REPORT DATE
December 1990

3. REPORT TYPE AND DATES COVERED
Final report

4. TITLE AND SUBTITLE
Distribution of PCB and Pesticide Contaminants in the Vicinity of the Times Beach Confined Disposal Facility, Buffalo, New York

5. FUNDING NUMBERS

6. AUTHOR(S)
Johannes M. Marquenie, John W. Simmers, Richard G. Rhett, Dennis L. Brandon

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
USAE Waterways Experiment Station, Environmental Laboratory, 3909 Halls Ferry Road, Vicksburg, MS 39180-6199

8. PERFORMING ORGANIZATION REPORT NUMBER
Miscellaneous Paper EL-90-24

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
USAE District, Buffalo, Buffalo, NY 14207-3199

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES
Available from National Technical Information Service, 5285 Port Royal Road, Springfield, VA 22161

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

This document reports the results of a biomonitoring study applied to quantify the role of the aquatic portion of the Times Beach confined disposal facility (CDF) at Buffalo, NY, as a potential source of contaminants for Lake Erie and the Niagara River. Mussels (*Elliptio dilatata*) were collected from a pristine lake and exposed as biomonitors in the Buffalo River, Lake Erie, and Times Beach. After 35 days exposure, the mussels were recovered and analyzed for polychlorinated biphenyls (PCBs), hexachlorobenzene, and pesticides (*op*-DDE and *pp*-DDE). The PCBs and pesticides were found to be present in the Buffalo River, Lake Erie, and the CDF. The biomonitor mussels placed inside the CDF accumulated PCB congeners 28, 44, 49, 52, 70, 87, and 101 to levels significantly higher than those of the mussels placed outside the dike. Concentrations of PCBs 138 and 153 were significantly higher in the biomonitors placed outside the dike. The pesticide concentrations in the biomonitor mussels were significantly higher in the CDF than in either the Buffalo River or Lake Erie.

14. SUBJECT TERMS
See reverse

15. NUMBER OF PAGES
37

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED		

SUMMARY

Mussels (*Elliptio dilatata*) were collected from a pristine lake and exposed as biomonitors in the Buffalo River, in Lake Erie, and at the Times Beach dredged material confined disposal facility (CDF). The mussels were allowed time to burrow either in the sediment or between stones for 35 days and were then collected and analyzed for polychlorinated biphenyls (PCBs), hexachlorobenzene and pesticides (*op*-DDE and *pp*-DDE).

After 35 days exposure, tissue analysis of the biomonitor mussels indicated that the Buffalo River contained bioavailable PCBs and pesticides in three areas: (a) the two main branches prior to the confluence forming the navigable portion, (b) the section flowing through the industrial area downstream of Cazenovia Creek, and (c) the mouth of the Buffalo River near the Buffalo Coast Guard Station.

Mussels exposed in the Times Beach CDF accumulated PCB congeners 28, 44, 49, 52, 70, 87, and 101 to levels above those of mussels exposed on the outside of the dike. The concentrations of higher chlorinated PCBs (138 and 153) were higher in biomonitors outside the dike. These data indicate that the same bioavailable PCBs were present on both sides of the porous dike but generally in significantly higher concentrations inside the dike. Therefore, the movement of PCB contaminants may be limited by the dike.

The same pesticide compounds were present in tissues of the biomonitor mussels placed in the Buffalo River, Lake Erie, and Times Beach. The results of the tissue analysis indicated that pesticide levels were significantly higher inside Times Beach as opposed to Lake Erie.

PREFACE

This report presents the results of research conducted jointly by the Technology for Society Division (MT) of The Netherlands Organization for Applied Scientific Research (TNO) and the US Army Engineer Waterways Experiment Station (WES), via cost-sharing contractual arrangements through the US Army Research, Development, and Standardization Group-United Kingdom (UK). The study was conducted for the US Army Engineer District, Buffalo, under the auspices of The United States/The Netherlands Memorandum of Understanding Concerning Dredging and Related Technology.

The report was prepared by Dr. Johannes M. Marquenie, MT-TNO, and by Dr. John W. Simmers and Messrs. Richard G. Rhett and Dennis L. Brandon, Ecosystem Research and Simulation Division (ERSD), Environmental Laboratory (EL), WES. Mr. D. K. Crawley and Dr. S. H. Kay of Delta State University, Cleveland, MS, and Dr. E. A. Stafford of the Rothamsted Experimental Station, Harpenden, UK, assisted in the conduct of the study and preparation of the report. Mr. J. C. Comati, Environmental Branch, US Army Research, Development, and Standardization Group-UK, was instrumental in establishing the cost-sharing research with TNO.

General supervision was provided by Dr. A. J. M. Schoot Uiterkamp, Head, Department of Biology, MT/TNO: Mr. Donald L. Robey, Chief, ERSD; and Dr. John Harrison, Chief, EL.

COL Larry B. Fulton, EN, was Commander and Director of WES. Dr. Robert W. Whalin was Technical Director. The Head Director of MT-TNO was Mr. I. J. C. Duyverman, and the Under Director at Delft was Dr. W. H. J. M. Wientjes.

This report should be cited as follows:

Marquenie, Johannes M., Simmers, John W., Rhett, Richard G., and Brandon, Dennis L. 1990. "Distribution of PCB and Pesticide Contaminants in the Vicinity of the Times Beach Confined Disposal Facility, Buffalo, New York," Miscellaneous Paper EL-90-24, US Army Engineer Waterways Experiment Station, Vicksburg, MS.

CONTENTS

DISTRIBUTION OF PCB AND PESTICIDE CONTAMINANTS IN THE VICINITY OF THE TIMES BEACH CONFINED DISPOSAL FACILITY, BUFFALO, NEW YORK

PART I: INTRODUCTION

Background

1. The Times Beach confined disposal facility (CDF), near Buffalo, NY, is a partially filled CDF that consists of upland, wetland, and aquatic habitats underlain by highly contaminated dredged material and confined by a porous rubble dike. The CDF is located at the mouth of the Buffalo River and is separated from Lake Erie by the dike. The US Army Engineer District, Buffalo, has been requested by State and Federal resource agencies to respond to the allegation that Times Beach is a source of contamination for adjacent Lake Erie and the nearby Niagara River.

2. Several contaminant mobility studies have been conducted for Times Beach (Kay, Simmers, and Marquenie 1986; Marquenie, Simmers, and Kay 1987; Stafford et al., in preparation). The information presented in these reports has concentrated mainly on the routes of contaminant movement in the upland and wetland ecosystems. The aquatic area of the site was studied to a limited extent by Kay, Simmers, and Marquenie (1986) and Marquenie, Simmers, and Kay (1987). Several fish species were collected from the Times Beach CDF and from a nearby reference area at the mouth of the Buffalo River (Kay, Simmers, and Marquenie 1986). Analysis of the fishes to determine concentrations of metals, polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs) indicated that the levels of copper, arsenic, and mercury were similar. Cadmium levels were higher in fishes from the Buffalo River, and PCB levels were higher in fishes collected from the CDF; however, these studies were preliminary in nature.

3. The mussel biomonitoring study described in this report assessed the presence of PCBs and pesticides in the Times Beach CDF aquatic area, the Buffalo River, and Lake Erie.

Objectives

4. The objectives of the study were as follows:

   a. To indicate the presence and distribution of bioavailable PCBs in the Buffalo River.

   b. To determine which bioavailable PCB congeners are present in the aquatic area of Times Beach.

   c. To compare the levels of PCB congeners in the tissues of biomonitors placed inside the CDF and in Lake Erie, i.e., movement of contaminants through the dike.

Approach

5. An aquatic biological monitoring system utilizes an organism sensitive to contaminants. Depending on the scope and objectives of the study, the criterion "sensitive" is defined and a suitable organism is selected. There are two alternative approaches, active and passive:

   a. Active biological monitoring (biomonitoring) uses a homogenous group of organisms either collected in the field or raised in the laboratory. The test organisms are divided at random into subgroups, and the subgroups are exposed in the field at different locations. These organisms are called "monitors."

   b. Passive biological monitoring uses organisms that occur naturally at the test site. There is a disadvantage in that the desired species may not always be present, numerous, or uniform in age. These organisms are called "indicators." Organisms are called indicators with reference to dynamic population reactions to contamination. Hueck (1976) further divided bioindicators into three classes:

      (1) Organisms with a negative response, which decrease in number.

      (2) Organisms with a neutral response, which maintain a static population.

      (3) Organisms with a positive response, which increase in number.

Whether species of a population decline, remain stable, or increase can be inferred from species lists and diversity indices.

6. There is another technique that does not place organisms in the field at all but takes the "field" to the organisms in the laboratory. This technique is generally described as a bioassay. Examples of the use of this approach by the US Army Engineer Waterways Experiment Station (WES) and The

Netherlands Organization for Applied Scientific Research (TNO) are given by Marquenie, de Kock, and Dinnen (1983).

7. Active biomonitoring, as used in this study, was developed using mussels in marine and estuarine environments (Goldberg 1976, Goldberg et al. 1978, de Kock and Kuiper 1981). Its application in the Dutch North Sea and estuaries has been described by de Kock (1983) and de Kock and Marquenie (1981). Active biomonitoring with freshwater mussels has been developed in The Netherlands by the Technology for Society Division, TNO, for The Netherlands Ministry of Transport and Public Works (Marquenie 1981, 1982). Development of the technique began in 1976; field verification took place in 1978. The technique has been used on many occasions in the Rhine River (from central Germany downstream) and the Meuse River (from northern France through Belgium into The Netherlands). Since 1982, it has been employed routinely at monthly intervals to monitor the water quality of the Rhine and Meuse Rivers.

8. A preferred organism for active biomonitoring in fresh water is the zebra mussel, *Dreissena polymorpha*, a mytillid species of surface waters that attaches to objects by byssus threads. Another suitable organism is a large unionid mussel, *Anodonta cygnea*, which can be used when it is desirable to collect organ-specific information on accumulation and elimination rates in field exposures. Both species are relatively tolerant to contaminants (Marquenie 1981) and at the same time capable of concentrating contaminants in their tissues, which are the general requirements set by Philips (1980a,b) for monitors.

9. Results of biomonitoring studies pertinent to the rationale of the present study have been published only in Dutch (Hueck 1976, del Castilho and Marquenie 1984). These results are summarized below:

a. Samples composed of the pooled tissues of 60 to 100 individual *D. polymorpha*, or 10 to 15 individual *A. cygnea*, have been found to reduce analytical variability to less than 10 percent.

b. Replicate exposures have demonstrated that the variability between duplicate samples of pooled individuals falls within the 10-percent analytical variation.

c. Variation in collection time (season) and place (field location) can cause extensive (orders of magnitude) differences in tissue concentrations.

These precepts indicate that, optimally, a monitoring test should replicate the locations that are typical for various microhabitats over time, rather than replicate locations at a single time. Furthermore, the animals should be exposed in the way most natural to the species. For this reason, *Dreissena* is exposed in cages in the water column and *Anodonta* is attached to cords and allowed to burrow in the sediment.

10. Concentrations of heavy metals and organic contaminants in the tissues of the animals appear to most closely reflect concentrations in the water and colloidal material and are not directly related to concentrations of the contaminants in suspended, filterable, particulate matter, or sediment (Baker, Capel, and Elsenreich 1986). An example of such a relationship is given for cadmium in Figure 1. An example of a concentration gradient of bioavailable cadmium downstream from a point source is shown in Figure 2. The importance of replication in time at a variety of locations is illustrated in Figure 3.

11. In the United States and Canada, *D. polymorpha* is not available; therefore, several alternative species of freshwater mussels were selected on the basis of local availability. The following genera are potentially useful as biomonitors in fresh water (depending on the region): *Lampsilis*, *Corbicula*, *Elliptio*, and *Anodonta*. This study utilized the freshwater mussel, *Elliptio dilatata*, a mussel occurring naturally near Times Beach and the Buffalo River.

PART II: MATERIALS AND METHODS

Selection of Contaminants for Analysis

12. Only PCB, hexachlorobenzene (HCB), and pesticide analyses were conducted during this study. These compounds do not occur naturally, are considered to be harmful to the environment, and are of concern in the Buffalo, NY, area. The PCBs existing as mixtures of distinct congeners have individual "fingerprints" that can facilitate the comparison of the PCB contaminants accumulated by the biomonitoring animals at several locations. In addressing the current issue of movement of contaminants through the Times Beach rubble dike, it was anticipated that the "fingerprints" of these mixtures would provide the most conclusive assessment of contaminant movement.

Selection of Biomonitoring Locations

13. The locations of biomonitoring animals are shown in Figures 4 and 5. The emphasis was to identify the existence of PCBs in the Buffalo River and to compare Times Beach to the immediately adjacent portion of Lake Erie (not to identify the sources of PCBs). In the Buffalo River, batches of mussels were exposed along the main channel and in all branches (Figure 4 and Table 1). At Times Beach, mussels were exposed along extensions of the transects previously established across the upland and wetland portions of the site and at regular intervals along the dike (Figure 5). Both the Buffalo River and the aquatic area of Times Beach have soft bottom material suitable for the biomonitoring animals. In Lake Erie there were no soft bottom sediments adjacent to Times Beach. The mussels were exposed at regular intervals along the lakeward side of the dike enclosing Times Beach and at two locations along the lakefront south of Times Beach. The breakwater lakeward of the Times Beach dike was inaccessible by boat due to storms at the time the study was initiated and could not be used as a biomonitoring station.

Collection of Mussels for Biomonitoring

14. Lake Chautauqua was found to be a suitable source of *E. dilatata*. The location was identified following discussions with the Department of

Biology, State University of New York, Fredonia, NY. The University supplied divers and a boat to collect the animals. The mussels were collected and sorted according to size; those selected for use in the study were moved to a spot in the same lake remote from human activities. The mussels were left there for 1 week prior to recollection for biomonitoring use.

Preparation of Mussels for Biomonitoring

15. The mussels were recollected 1 day before field exposure, taken to Buffalo, and individually attached (using "5-minute" epoxy adhesive) to 50-cm-long strands of nylon cord. The mussels were then sorted into batches of 15 randomly selected individuals. The following morning, one batch was taken to each biomonitoring site, attached to a sufficiently long cord, and sunk to the bottom where the mussels were allowed to burrow into the sediment. The end of the cord was fastened to the nearest immovable object.

16. The mussels were recollected 35 days later and frozen on dry ice (-60° C) without depurating. They were then shipped to TNO Laboratories in The Netherlands and stored at -18° C until analyzed.

Chemical Analysis

17. In the TNO laboratory, the soft tissues of the mussels were dissected free of the shells with titanium scalpels and placed in acid-washed glassware. The tissues of the mussels from each batch were pooled, their wet weight recorded, and the total sample homogenized using an "Ultra-turrax" homogenizer (TNO-manufactured) equipped with titanium blades. Subsamples of the resulting homogenate were taken for the appropriate chemical analysis and for determination of dry and ash weights. The remaining tissue homogenate was stored frozen (-18° C) for any future analysis of heavy metal or organic contaminants.

18. Concentrations of PCB congeners, *op*-DDE, *pp*-DDE, and HCB were determined by capillary gas chromatography after enzymatic treatment of the homogenate, extraction, and cleanup (Marquenie, Simmers, and Kay 1987). Ash-free dry weight was determined gravimetrically by weighing, drying at 105° C (16 hr), ashing at 600° C (24 hr), and reweighing. For convenience, the International Union of Pure and Applied Chemists (IUPAC) numbers were used to

identify PCB congeners. A key to the IUPAC nomenclature is provided as Table 2.

PART III: RESULTS AND DISCUSSION

Exposure of Mussels

19. The biomonitoring locations and the numbers of mussels retrieved and analyzed after 35 days exposure are listed in Table 3. Also given are the total wet weight of the soft tissues retrieved and the calculated average wet tissue weight per mussel. Some mussels were lost from nearly all the batches of 15 mussels set out. This was attributed to poor adhesion, vandalism, or heavy storm action. Some complete batches of mussels were lost. Biomonitoring efforts by the Dutch also resulted in initial losses, but these losses were reduced significantly in subsequent efforts as techniques appropriate to each waterway were developed, as is now possible at Buffalo.

20. The percentage of ash-free dry tissues, dry tissues, and calculated average weights of the ash-free dry tissues per animal are listed in Table 4. These data show considerable variation in the percentage of dry tissues, and consequently, in the moisture content of the mussel homogenate. This is due in part to water loss during handling and in part to the internal osmotic status of the animals. The analytical results have been converted to an ash-free dry weight basis to remove the contribution of noncombustible material in determining contaminant concentrations. Therefore, concentrations of contaminants have been expressed in terms of ash-free dry weight to avoid bias from salts in the tissue. (Concentrations on a wet weight basis that relate to regulatory criteria are given in Table 5.)

Concentrations of PCBs

21. Table 6 lists the concentrations of PCB congeners in mussel tissues. Occasionally, a single peak in a chromatogram could not be properly quantified. It is for this reason that data for PCB 180 in the Buffalo River (Station 3) and for PCB 138 at the Coast Guard Station (Station 1) are questionable.

Buffalo River

22. The data presented in Table 6 show that the Buffalo River is contaminated with PCBs. Concentrations of all congeners, except 87, were found in the tissues of the mussels exposed upstream (Stations 12 and 13), beyond

the navigable stretch (Figure 4). Concentrations in mussels exposed in the stretch crossing under the highway at Station 12 (Cayuga Creek, Table 6) appeared to be generally higher than those in mussels from the stretches along School Road in Cazenovia Creek (Station 13). In both of these stretches, the current was strong and the water shallow and clear.

23. Below the junction of the two tributaries (Cayuga Creek and Buffalo Creek), the river widens and deepens, and the current is slower (Figure 4). Here the water becomes turbid from algae and suspended organic matter. These conditions may strongly reduce the bioavailability of PCBs to mussels, as is shown by the PCB concentrations found in the biomonitors at the two locations farther downstream (Stations 9 and 10). At the bridge, immediately upstream from Cazenovia Creek (Station 8), some PCB congener concentrations tended to be higher than at Stations 9 and 10 farther upstream. Cazenovia Creek (Station 7) apparently contributes little to the PCB contamination of the Buffalo River as measured by biomonitor mussels.

24. An increase in PCB concentrations was found in mussels in the river at Station 6. This suggests that PCBs may have entered the Buffalo River at this point. The ratio between the different congeners suggests that the area is contaminated with a mixture of PCBs in which higher chlorinated compounds (PCBs 138, 153, and 180) predominate (Table 6). The concomitant finding, that DDE concentrations at this station are also elevated, leads to two suspected sources of contamination: surface runoff water and industrial wastewater.

25. Farther downstream at Stations 3, 4, and 5 (Figure 5), the PCB concentrations in the mussels are lower. This may be due to reduced bioavailability caused by adsorption of PCBs on suspended matter or sediments. The PCB concentrations in the mussels rise again at the mouth of the Buffalo River (Station 1).

26. These results are comparable to those of a freshwater mussel biomonitoring study in the Meuse River (The Netherlands) to the extent that bioavailability was found to decrease downstream from potential sources (Marquenie, Simmers, and Kay 1985).

27. In summary, elevated PCB concentrations occurred in mussels exposed in three stretches of the Buffalo River:

- a. The two main tributaries that feed into the navigable portion.
- b. The industrial area contaminated mainly with higher chlorinated PCBs.
- c. The mouth of the Buffalo River.

If the sources of the PCBs are of concern, further studies are required. The ratios of PCB congeners in mussel tissues from the Meuse River were used to reveal specific sources. In the Buffalo River, however, the composition of PCB mixtures has not been used to reveal specific sources but could be used after additional studies. The monitoring can be confined to the river areas found to be most heavily contaminated with bioavailable PCBs.

Lake Erie

28. Mussels exposed in Lake Erie showed a variation in PCB concentrations. The tissue concentrations of PCBs appear to generally increase from Station 22 to 14 (Table 6). The contamination of this part of Lake Erie (Buffalo Harbor) apparently is not uniform. However, the area covered in this study was limited, and the true background concentrations of PCBs in mussels in Lake Erie are not known. The mussels exposed in Lake Erie contained all PCB congeners (except PCB 87) above the detection limit.

Times Beach

29. Mussels exposed in the CDF contained very low concentrations of the higher chlorinated PCBs (138, 153, and 180) in comparison to those in Lake Erie, even though PCBs 138 and 153 had been detected in the dredged material in previous studies (Figure 6 and Table 7). The mussels exposed inside the Times Beach CDF contained high concentrations of lower chlorinated PCBs (28, 44, 49, 70, 87, and 101). Moreover, PCB 87, not detected in mussels from Lake Erie, occurred in all mussels from Times Beach (Figure 6). The results summarized in Figure 6 indicate that concentrations of PCBs (except PCB 28) in the dredged material were higher than the concentrations in the mussels inside Times Beach. In addition, PCB concentrations in the mussels placed outside the dike were generally lower than the PCB concentrations in the mussels inside the dike (Figure 6). The mussels exposed at Times Beach showed some variation in PCB uptake (Tables 6 and 8). The source of this variation may be discovered through the analysis of the dredged material from the aquatic area and through laboratory bioassay procedures.

30. It is of note that the highest PCB concentrations were found in a sample of mussels suspended by a float above the sediment at Station D8 (Table 6). The tissue levels from the Station D8 float were not included in the statistical comparison of the data. The elevated PCB levels of the Station D8 float (Table 6) may be the result of the mussels processing mainly water and associated unfilterable colloidal particles rather than filterable particulate material (Baker, Capel, and Elsenreich 1986).

31. To determine if the mixtures of PCB congeners accumulated inside and outside the site were different, statistical tests were performed. The mussel data from the CDF were evaluated using a multivariate analysis of variance procedure (MANOVA). The data set contained 12 observations (eight inside the site and four outside the site) and 11 PCB congeners. Attempts to evaluate the data using all congeners were impractical. Since correlations between many of the congeners were high, the data were grouped on that basis; however, other combinations were considered. Group 1 consisted of congeners 44, 49, 52, 70, 87, and 101. The remaining congeners (15, 28, 138, 153, and 180) made up Group 2.

32. Levene's test of homogeneity of variance was applied to the data for Group 1 (Snedecor and Cochran 1980). This assumption was rejected. The application of Levene's test to square root (SQRT) transformed data failed to reject the null hypothesis. This was true for every congener in Group 1 except 87. Group 1 congeners 44, 49, 52, 70, and 101 were then evaluated using the MANOVA. Using Wilk's criterion (Morrison 1976), the null hypothesis of equal means was rejected. Therefore, the mussel tissue concentrations of the congeners inside the disposal area are statistically different from those outside the disposal area. Roy-Bose Simultaneous Confidence Intervals were used to determine individual differences. A 95-percent confidence interval was calculated for each congener. The mean tissue concentration of mussels placed outside the dike was subtracted from the concentration of mussels placed inside the CDF (i.e., mean inside - mean outside). When the mean concentrations are higher inside, this expression is greater than zero. Table 9 lists the confidence intervals for each congener in Group 1, except 87. Congeners 49, 52, and 70 have lower confidence limits that are greater than zero. This is evidence that the mean tissue concentration inside the CDF is greater than the mean tissue concentration outside the CDF in adjacent Lake Erie. The Wilk's Criterion and Roy-Bose Simultaneous Confidence Intervals were used as described in Morrison (1976). Congener 87 is consistent with this result as tissue concentrations inside the disposal area are at least 20 times higher than those outside the disposal area (Table 8).

33. Group 2 congeners were also evaluated using a MANOVA. This group consisted of congeners 15, 28, 138, 153, and 180. A SQRT transformation (15, 28, 180) and 10× common log (138, 153) were used to achieve homogeneity of variance. The null hypothesis of equal means was rejected. However, the Roy-Bose procedure failed to detect any individual differences. Tissue

concentrations of each congener were also compared using an analysis of variance (ANOVA). The transformations noted above were used to achieve a homogeneous variance. A nonparametric procedure was used to evaluate congener 87. Table 8 lists the results of these procedures. All differences identified by the MANOVA were also identified by the ANOVA. In addition to congeners 49, 52, and 70, statistical differences were found between mean tissue concentrations of congeners 28, 44, 87, 101, 138, and 153.

34. Congeners 15 and 180 appear to be present in statistically similar levels in the tissues of mussels placed on either side of the CDF dike (Table 8). These congeners are also widely distributed in the Buffalo River (Table 6). There are several possible explanations for the widespread presence of PCB 15. This dichlorobiphenyl may have been present in high concentrations in the initial contamination of the area, as a result of aerial deposition, or as suggested by Brown et al. (1984), as a by-product of microbial dechlorination of more highly chlorinated PCB congeners. In the case of PCB 180, Smith et al. (1985) consider this congener to be "recalcitrant" or very resistant to dechlorination. If this is true, PCB 180 may be persistent in its original form while being widely dispersed from the original point source.

35. In summary, the data presented in Tables 7-9 and Figure 6 indicate that the concentrations of PCB congeners in biomonitor mussels inside and outside the porous Times Beach dike are different and that higher concentrations of the lower chlorinated PCBs were generally present in the mussels from inside the dike. Congener 87 was present in dredged material and biomonitor mussels in the CDF but was below detection limits in Lake Erie outside the dike. This finding, in conjunction with the significantly different concentrations inside and outside the CDF, suggests that PCB movement may be limited by the dike. However, these data can neither substantiate nor refute the position that Times Beach is a source of PCB contamination of Lake Erie. If the PCB congeners in question are associated with a colloidal phase in the water column, as suggested by Baker, Capel, and Elsenreich (1986), it is possible that some PCB contaminants move with water through the dike as lake stages change, and are diluted in Lake Erie.

Concentrations of Pesticides

36. Concentrations of pesticides in mussels are given in Table 10 (µg/kg ash-free dry weight) and in Table 5 (wet weight). Like PCBs, pesticides were present far upstream in the Buffalo River system before the two main tributaries merge (Stations 12 and 13) and increased in tissue levels downstream with the highest concentration at Station 1.

37. Pesticide concentrations in biomonitoring mussels from Lake Erie are slightly lower than the highest Buffalo River locations. The concentrations in Lake Erie at Station 22 were below detectable limits for DDE. The levels of pesticides accumulated by mussels placed in the Times Beach aquatic area were significantly higher than the levels in the mussels placed outside in Lake Erie (Table 11). As discussed for PCBs, these data suggest that pesticide movement may be limited by the dike but are inconclusive regarding Times Beach as a source of pesticide contamination for Lake Erie.

Comparison of Times Beach to Dutch Experiences

38. The mussels exposed at the Times Beach CDF accumulated PCBs and pesticides. In food-chain studies conducted in The Netherlands, diving ducks were fed contaminated mussels (*D. polymorpha*) from a contaminated watershed for 3 years. As a result, the ducks sometimes suffered breeding failure and mortality of embryos. The suspected causes were related to PCBs, DDE, and mercury contamination in the food. The contaminant concentrations in the freshwater mussels fed to the ducks are given in Table 12. While the concentrations of contaminants in mussels exposed at Times Beach were generally lower than those from the contaminated watershed (Haringvliet) suspected to be responsible for harmful effects in the ducks, they were higher than the levels of the uncontaminated reference (Markermeer). The PCB levels at the Times Beach CDF may have harmful effects on waterfowl.

PART IV: CONCLUSIONS

Buffalo River

39. Stretches of the Buffalo River appeared to be contaminated with PCBs and pesticides, as shown by biomonitors.

40. Some contamination was present upstream of the navigable stretch. Of the sections of the river studied, the industrial section appears to be the most severely contaminated.

Times Beach

41. The mussels exposed in Times Beach were found to contain higher concentrations of the lower chlorinated PCBs and pesticides than mussels exposed on the Lake Erie side of the dike, but the same bioavailable PCBs were present on both sides of the porous dike with the exception of PCB 87.

42. Congener 87 was present in both the dredged material and biomonitor mussels inside the dike but was undetected in mussels outside the dike. This suggests that one or possibly two PCB congeners were contained by the dike.

43. Congeners 138 and 153 were present in the Times Beach dredged material but were not detected in the biomonitoring mussels inside the dike. These PCB congeners were present in the biomonitors placed outside the dike. The source of PCB 138 and 153 to the biomonitor mussels outside the dike may have been from origins other than the CDF.

44. Although tissue concentrations of PCBs from the biomonitors were statistically different inside and outside the dike, this information neither supports nor refutes the position that Times Beach contributes to the PCB contamination of Lake Erie and the Niagara River. The presence of the same PCB congeners in biomonitoring animals on both sides of the dike suggests the need for further evaluation of the ambient levels of PCBs in Lake Erie to determine the origins of the PCB congeners detected in the biomonitors.

45. The total PCB concentrations found in mussels exposed within Times Beach were elevated in comparison to adjacent Lake Erie and may be of ecotoxicological significance. Further studies at Times Beach should consider the possible effects of organic contaminants on animals in contact with the dredged material, including waterfowl, muskrat, and beaver.

46. The pesticides *op*-DDE and *pp*-DDE and hexachlorobenzene are present in both Times Beach and Lake Erie. The significantly higher levels in Times Beach indicate that the dike may limit the pesticide movement.

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Table 1

Detailed Location Descriptions of Biomonitoring Stations in the Buffalo River*

Station	Location
1	Across the Buffalo River from Buoy 4 at the Coast Guard Station boat dock
2	"Sloop number 7" sign at the commercial small boat dock across the Buffalo River from the Buffalo Naval Park
3	Pilings near the skyway in the ship channel
4	Pilings under the skyway
5	The right (west) bank of the Buffalo River attached to pilings at the Cargill grain elevators
6	The right bank directly across the Buffalo River from Allied Chemical
7	Beneath the Bailey Avenue bridge crossing Cazenovia creek
8	Beneath the Bailey Avenue bridge crossing the Buffalo River
9	Approximately 100 m upstream of the Seneca Street bridge
10	Beneath the Harlem Road bridge at the junction of Cayuga and Buffalo Creeks
11	Beneath the Clinton Street bridge in Cayuga Creek
12	Downstream of the Clinton Street bridge in Buffalo Creek
13	Across the creek from School Road

* See Figure 4.

Table 2

Key to Polychlorinated Biphenyl Nomenclature Designated by IUPAC Number

PCB (IUPAC No.)	PCB Congener	
15	4,4'	Dichlorobiphenyl
28	2,4,4'	Trichlorobiphenyl
44	2,2',3,5'	Tetrachlorobiphenyl
49	2,2',4,5'	Tetrachlorobiphenyl
52	2,2',5,5'	Tetrachlorobiphenyl
70	2,3',4',5	Tetrachlorobiphenyl
87	2,2',3,4,5'	Pentachlorobiphenyl
101	2,2',4,5,5'	Pentachlorobiphenyl
138	2,2',3,4,4',5'	Hexachlorobiphenyl
153	2,2',4,4',5,5'	Hexachlorobiphenyl
180	2,2',3,4,4',5,5'	Heptachlorobiphenyl

Table 3

Exposure Sites, Number of Animals (*Elliptio dilatata*) Retrieved (Out of 15), Total Wet Weight per Sample, and Average Wet Weight per Individual

	Station Location*	Number of Animals	Wet Weight, g	Average Wet Weight, g
		Buffalo River		
1	Coast Guard Station	8	140.14	17.51
2	Marina	Lost		
3	Pilings across from Times Beach	15	225.18	15.01
4	Pilings under skyway	11	215.85	19.62
5	Pilings at grain elevator	12	185.34	15.44
6	Across from Allied Chemical	15	317.40	21.16
7	Cazenovia Creek	10	202.78	20.27
8	Bridge upstream from Cazenovia Creek	11	206.48	28.77
9	Past Grassy Island	9	143.92	15.90
10	Channel across from bridge	14	170.90	12.20
11	End of navigable stretch	Lost		
12	Under highway bridge	13	232.92	17.91
13	School Road	9	139.92	15.54
		Lake Erie Outside Times Beach		
14		2	29.04	14.52
14A		Lost		
15		Lost		
16		Lost		
17		Lost		
18		Lost		
19		13	173.72	13.36
20		10	164.88	16.48
21		Lost		
22		4	38.29	9.57
		Inside Times Beach		
23		Lost		
24		8	152.80	19.10
25		7	115.80	16.54
		Transect Locations		
D6		10	122.54	12.25
D7		8	118.76	14.84
D8		12	205.01	17.08
D8	(float)	2	41.57	20.78
E7		9	159.94	17.77
E8		9	213.72	23.74
E9		7	104.57	14.93

* See Figures 4 and 5.

Table 4

Percentage of Ash-Free Dry Material, Percentage of Dry Material in Each Sample, and Average Ash-Free Dry Weight per Individual Mussel (*Elliptio dilatata*) in Each Sample

Station Location*	Ash-Free Dry Weight percent	Dry Weight percent	Ash-Free Dry Weight, g (average)
Buffalo River			
1 Coast Guard station	8.59	10.89	1.50
3 Pilings across Times Beach	8.07	10.85	1.21
4 Pilings under skyway	9.03	11.64	1.77
5 Pilings at grain elevator	6.68	9.25	1.03
6 Across from Allied Chemical	7.87	9.79	1.66
7 Cazenovia Creek	7.70	10.05	1.56
8 Bridge upstream from Cazenovia Creek	9.00	11.43	1.68
9 Past Grassy Island	6.67	8.90	1.06
10 Channel across from bridge	5.92	8.25	0.72
12 Under highway bridge	5.72	7.71	1.02
13 School Road	4.58	5.77	0.71
Lake Erie Outside Times Beach			
14	9.73	12.14	1.41
19	7.68	9.70	1.02
20	5.05	6.85	0.83
22	6.52	9.01	0.62
Inside Times Beach			
24	6.54	8.29	1.25
25	9.56	11.55	1.58
Transect Locations			
D6	5.68	7.77	0.69
D7	6.25	8.08	0.92
D8	6.18	8.41	1.05
D8 (float)	9.55	11.72	1.98
E7	7.07	8.49	1.25
E8	3.83	4.94	0.91
E9	8.10	10.21	1.21

* See Figures 4 and 5.

Table 5

Concentrations of PCBs and Pesticides in the Soft Tissues of Elliptio dilatata

(μg/kg wet weight)

	PCB (IUPAC No.)											Pesticide		
Station Location*	15	28	52	49	44	70	101	87	153	138	180	op-DDE	pp-DDE	HCB
Buffalo River														
1 Coast Guard Station	5.7	3.9	5.1	3.2	7.7	9.1	5.2	2.6	7.3	7.3	4.3	1.4	4.2	1.4
3 Pilings across Times Beach	4.9	0.4	2.7	2.0	<0.1	3.5	<0.1	<0.1	<0.1	<0.1	53.0	0.9	2.3	<0.1
4 Pilings under skyway	5.5	1.5	3.8	2.8	<0.1	5.0	<0.1	1.0	<0.1	<0.1	1.3	1.1	1.7	<0.1
5 Pilings at grain elevator	6.0	1.6	3.6	2.7	<0.1	4.5	<0.1	<0.1	<0.1	<0.1	1.3	0.9	1.0	0.2
6 Across from Allied Chemical	8.1	2.4	4.2	5.4	9.0	7.2	4.9	1.6	9.6	9.4	7.3	1.2	2.5	1.6
7 Cazenovia Creek	4.7	<0.1	0.7	<0.1	<0.1	<0.1	<0.1	0.5	<0.1	<0.1	0.2	<0.1	0.4	0.8
8 Bridge upstream from Cazenovia Creek	5.7	<0.1	1.5	1.1	<0.1	4.2	<0.1	<0.1	<0.1	<0.1	3.6	<0.1	0.9	1.2
9 Past Grassy Island	6.0	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.5	<0.1	<0.1	3.2	<0.1	0.7	<0.1
10 Channel across from bridge	8.5	<0.1	0.5	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.3	0.6
12 Under highway bridge	5.2	0.7	1.3	2.2	7.6	2.4	1.3	<0.1	1.8	2.7	1.1	0.5	1.5	0.4
13 School Road	6.5	0.6	1.4	0.9	7.4	1.4	0.4	<0.1	1.0	2.1	0.6	0.3	1.0	0.3
Lake Erie Outside Times Beach														
14	7.6	2.0	4.5	1.3	15.0	5.7	2.6	<0.1	4.1	5.6	2.4	0.8	3.2	0.8
19	6.1	1.1	2.8	2.9	9.4	3.1	0.7	<0.1	2.8	3.7	1.5	0.5	1.9	0.8
20	11.0	0.5	1.4	1.8	8.5	1.8	0.4	<0.1	1.7	2.3	0.9	0.3	1.1	0.5
22	7.9	<0.1	0.8	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.6
Inside Times Beach														
24	4.1	5.1	14.0	10.0	4.7	14.0	3.4	2.8	<0.1	<0.1	<0.1	1.8	6.8	2.4
25	6.4	13.0	21.0	15.0	10.0	19.0	5.8	4.2	<0.1	<0.1	0.8	5.8	5.5	1.7
Transect Locations														
D6	9.3	25.0	37.0	27.0	23.0	32.0	7.8	5.7	<0.1	<0.1	<0.1	8.2	7.6	2.4
D7	5.1	16.0	27.0	17.0	13.0	21.0	3.4	3.6	<0.1	<0.1	<0.1	5.9	4.9	1.6
D8	6.8	14.0	24.0	18.0	12.0	25.0	7.8	5.3	<0.1	<0.1	0.7	2.5	6.6	2.2
D8 (float)	3.6	15.0	29.0	22.0	45.0	30.0	11.0	6.5	<0.1	<0.1	0.6	3.5	8.2	2.6
E7	7.2	21.0	31.0	24.0	19.0	30.0	8.4	5.6	<0.1	<0.1	0.3	2.4	6.8	2.4
E8	6.8	18.0	29.0	22.0	16.0	31.0	9.3	6.2	<0.1	<0.1	0.5	4.6	7.8	2.5
E9	5.0	18.0	31.0	23.0	16.0	32.0	11.0	6.3	<0.1	<0.1	0.9	2.1	7.9	2.7

* See Figures 4 and 5.

Table 6

Concentrations of PCB Congeners in *Elliptio dilatata*

($\mu g/kg$ ash-free dry weight)

Station Location*	PCB Congener (IUPAC No.) 15	28	52	49	44	70	101	87	153	138	180
Buffalo River											
1 Coast Guard station	66.3	45.4	59.3	37.2	89.6	105.9	60.5	30.2	84.9	1.5	50.0
3 Pilings across Times Beach	39.5	3.2	21.7	16.1	<0.8	28.2	<0.8	<0.8	<0.8	<0.8	656.7
4 Pilings under skyway	49.6	13.5	34.3	25.2	<0.9	45.1	<0.9	9.0	<0.9	<0.9	14.3
5 Pilings at grain elevator	40.0	10.6	24.0	18.0	<0.6	30.0	<0.6	6.0	<0.6	<0.6	19.4
6 Across from Allied Chemical	63.7	18.8	33.0	42.4	70.8	56.6	38.5	12.5	75.5	73.9	92.7
7 Cazenovia Creek	36.1	<0.7	5.3	<0.7	<0.7	<0.7	<0.7	3.8	<0.7	<0.7	2.5
8 Bridge upstream from Cazenovia Creek	51.3	<0.9	13.5	9.9	0.9	37.8	<0.9	7.2	<0.9	<0.9	39.9
9 Past Grassy Island	40.0	<0.6	<0.6	<0.6	<0.6	<0.6	<0.6	3.3	<0.6	<0.6	47.9
10 Channel across from bridge	50.3	<0.5	2.9	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<1.6
12 Under highway bridge	29.7	4.0	7.4	12.5	43.4	13.7	7.4	<0.5	10.2	15.4	19.2
13 School Road	29.7	2.7	6.4	4.1	33.8	6.4	1.8	<0.4	4.5	9.6	13.1
Lake Erie Outside Times Beach											
14	73.9	19.4	43.7	12.6	145.9	55.4	25.2	<0.9	39.8	54.4	24.6
19	46.8	8.4	21.5	22.2	72.1	23.8	5.3	<0.7	21.5	28.4	19.5
20	55.5	2.5	7.0	9.0	42.9	9.0	2.0	<0.5	8.5	11.6	17.8
22	51.5	<0.6	5.2	<0.6	<0.6	<0.6	<0.6	<0.6	<0.6	<0.6	<1.5
Inside Times Beach											
24	26.8	33.3	91.5	65.5	30.7	91.5	22.2	18.3	<0.6	<0.6	<1.5
25	61.1	124.2	200.7	143.4	95.6	181.6	55.4	40.1	<0.9	<0.9	8.3
Transect Locations											
D6	52.8	142.0	210.1	153.3	130.6	181.7	44.3	32.3	<0.5	<0.5	<1.7
D7	31.8	100.0	143.7	106.2	81.2	131.2	21.2	22.5	<0.6	<0.6	<1.6
D8	42.0	86.5	148.3	111.2	74.1	154.5	48.2	32.7	<0.6	<0.6	11.3
D8 (float)	34.3	143.2	276.9	210.1	429.7	286.5	105.0	62.0	<0.9	<0.9	6.2
E7	50.9	148.4	219.1	169.6	134.3	212.1	59.3	39.5	<0.7	<0.7	4.2
E8	26.0	68.9	111.0	84.2	61.2	118.7	35.6	23.7	<0.3	<0.3	13.0
E9	40.5	145.8	251.1	186.3	129.6	259.2	89.1	51.0	<0.8	<0.8	11.1

* See Figures 4 and 5.

Table 7

PCB Concentrations in Wetland Surface Dredged Material

(µg/kg dry weight)

PCB (IUPAC No.)	Individual Collections*						Mean ± SD
28	94	84	130	54	55	64	80.2 ± 29.2
49	170	190	160	110	150	220	166.7 ± 37.2
52	230	290	220	180	220	340	246.7 ± 57.8
70	220	230	210	150	120	290	203.3 ± 60.5
87	59	89	76	48	100	120	82.0 ± 26.6
101	89	130	110	71	140	190	255.2 ± 313.4
138	40	79	41	51	71	110	65.3 ± 27.0
153	26	48	42	23	71	79	48.2 ± 22.9

* Data from Marquenie, Simmers, and Kay 1987.

Table 8

Concentrations of PCB Congeners in Elliptio dilatata Inside and Outside the Times Beach Dike (µg/kg ash-free dry weight)

PCB (IUPAC No.)	Inside	Outside
15	41.49 ± 12.83A*	56.92 ± 11.86A
28	106.14 ± 41.55A	7.72 ± 8.46B
44	92.16 ± 37.51A	65.36 ± 61.18B
49	127.45 ± 42.42A	11.10 ± 8.95B
52	171.94 ± 56.46A	19.35 ± 17.80B
70	166.31 ± 54.00A	22.20 ± 24.12B
87	32.51 ± 10.87A**	0.67 ± 0.17B
101	46.91 ± 22.08A	8.27 ± 11.45B
138	0.62 ± 0.18B	23.75 ± 23.41A
153	0.62 ± 0.18B	17.60 ± 17.12A
180	6.59 ± 4.88A	15.85 ± 9.99A

* Means followed by the same letter in each row are not different at the level of significance as determined using an ANOVA.

** Wilcoxon two-sample test used at the 0.05 level of significance.

Table 9

95-Percent Simultaneous Confidence Intervals for Group 1 PCB Congeners

PCB (IUPAC No.)		Confidence Interval Ui1*	Confidence Interval Ui2**	
52	<0.91	U11	U12	<16.89
49	<1.22	U21	U22	<14.98
44	<-9.2	U31	U32	<13.6
70	<0.01	U41	U42	<16.39
101	<-2.35	U51	U52	<10.15

* Ui1, mussels inside the disposal area.

** Ui2, mussels outside the disposal area.

Table 10

Concentrations of Pesticides in *Elliptio dilatata*

(μg/kg ash-free dry weight)

Station Location*	*op*-DDE	*pp*-DDE	HCB
Buffalo River			
1 Coast Guard station	16.2	48.8	16.2
3 Pilings across Times Beach	11.1	28.5	<1.2
4 Pilings under skyway	12.1	18.8	<1.2
5 Pilings at grain elevator	13.4	14.9	2.9
6 Across from Allied Chemical	15.2	31.7	20.3
7 Cazenovia Creek	<1.2	5.1	10.3
8 Bridge upstream from Cazenovia Creek	<1.1	9.9	13.3
9 Past Grassy Island	<1.4	10.4	<1.4
10 Channel across from bridge	<1.6	5.0	10.1
12 Under highway bridge	8.7	26.2	6.9
13 School Road	6.5	21.8	6.5
Lake Erie			
14	8.2	32.8	8.2
19	6.5	24.7	10.4
20	5.9	21.7	9.9
22	<1.5	<1.5	9.2
Times Beach			
24	27.5	103.9	36.6
25	60.6	57.5	17.7
Transect Locations			
D6	144.3	133.8	42.2
D7	94.4	78.4	25.6
D8	40.4	106.7	35.5
D8 (float)	36.6	85.8	27.2
E7	33.9	96.1	33.9
E8	120.1	203.6	65.3
E9	25.9	97.5	33.3

* See Figures 4 and 5.

Table 11

Concentrations of Pesticides in *Elliptio dilatata* Inside and Outside the Times Beach Dike (μg/kg ash-free dry weight)

Pesticide	Inside	Outside
op-DDE	68.39 ± 42.75A*	5.53 ± 2.47B
pp-DDE	109.69 ± 41.05A	20.18 ± 11.52B
HCB	36.26 ± 13.00A	9.43 ± 0.83B

* Means followed by the same letter in each row are not different at the 0.05 level of significance as determined by an ANOVA.

Table 12

Concentrations of PCBs, Pesticides, and PAHs in the Freshwater Bivalve *Dreissena polymorpha*

Contaminant	Haringvliet		Markermeer	
PCB (IUPAC No.)				
15	1,573	1,425	173	72
28	219	193	43	20
52	397	347	<1	<1
49	287	257	<1	<1
44	602	488	173	190
70	315	411	<1	22
101	506	499	41	24
87	100	83	19	5
153	575	513	47	30
138	205	180	16	13
Pesticides				
op-DDE	246	231	<1	<1
pp-DDE	82	231	15	8
HCB	126	114	36	24
PAHs				
Phenanthrene	7	58	57	10
Anthracene	15	15	4	1
Fluoranthene	547	578	161	66
Pyrene	534	526	90	<51
3,6-dimethyl-phenanthrene	<27	<26	<25	<51
Triphenylene	492	436	68	<51
Benzo(b)fluorene	<41	<39	<37	<38
Benzo(a)anthracene	383	334	28	<13
Chrysene	465	411	64	<38
Benzo(e)pyrene	260	205	<74	<76
Benzo(j)fluoranthene	<68	<64	<62	<63
Pyrene	37	33	19	9
Benzo(b)fluoranthene	315	282	42	20
Benzo(k)fluoranthene	150	141	20	10
Benzo(a)pyrene	192	167	14	7
Dibenzo(a,j)anthracene	<41	<39	<37	<25
Dibenzo(a,i)pyrene	<27	<26	<25	<25
Benzo(g,h,i)perylene	178	154	<62	<63
Dibenzo(a,h)anthracene	<96	<90	<87	<89
Indeno(1,2,3-c,d)-pyrene	97	86	20	10
3-methylcholanthrene	<7	<6	<6	<6
Antranthrene	7	7	<4	<5

Note: The mussels were collected from the two freshwater basins (Haringvliet (polluted) and Markermeer (reference)) and fed to ducks for a period of 3 years. Concentrations are based on ash-free dry weights (μg/kg).

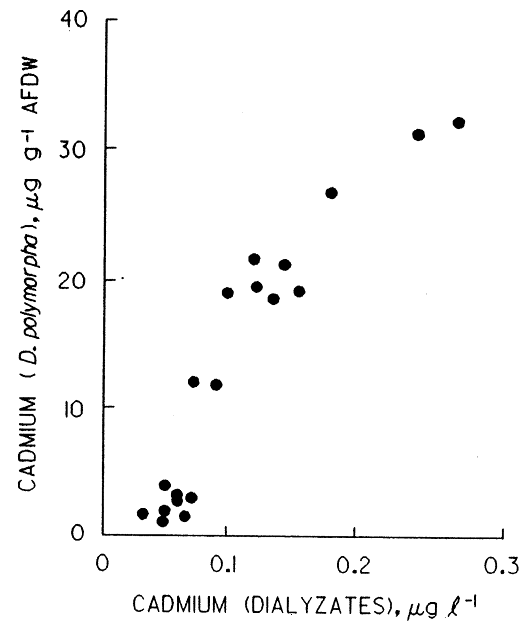

Figure 1. Concentrations of cadmium in *D. polymorpha* (ash-free dry weight) after 40 days of exposure at various locations in two rivers compared with concentrations of "free" cadmium in collected dialyzates in situ

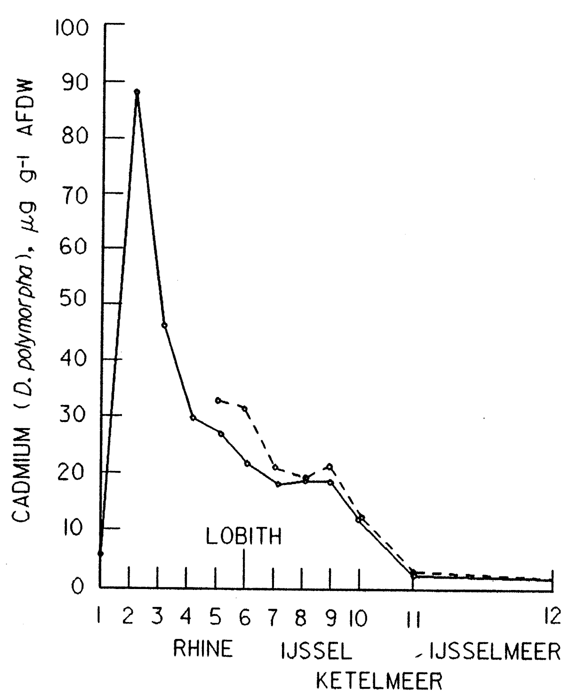

Figure 2. Concentrations of cadmium in soft tissues of *D. polymorpha* (ash-free dry weight) at various locations in the Rhine and its downstream lakes in 1982. (The Rhine enters The Netherlands at Lobith)

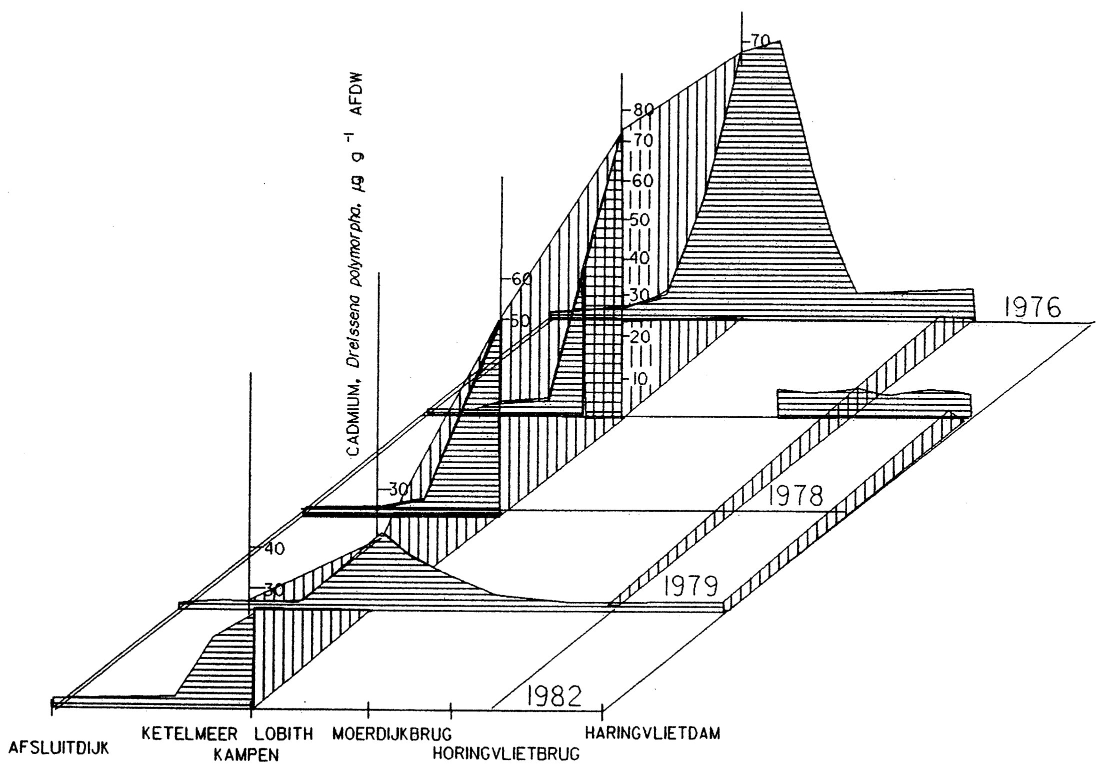

Figure 3. Bioavailability of cadmium in Dutch inland surface waters. Concentrations in *D. polymorpha* (ash-free dry weight) in successive years (1976-82) at different locations. From Lobith, where the Rhine enters The Netherlands, the two main stretches are followed: the river Waal, feeding the Haringvliet basin, and the river IJssel, feeding the IJsselmeer

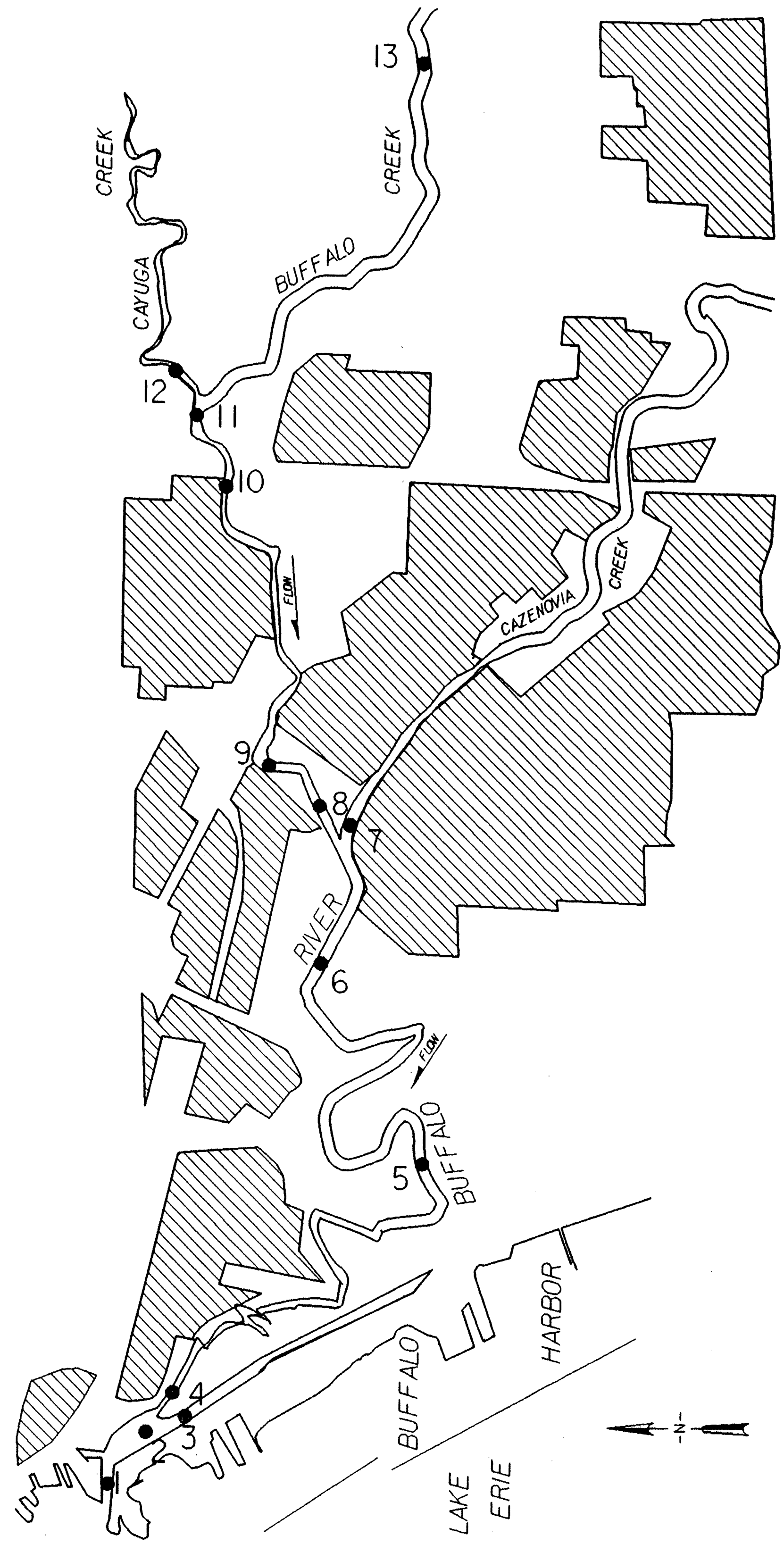

Figure 4. Biomonitoring locations in the Buffalo River
(see Tables 1 and 3)

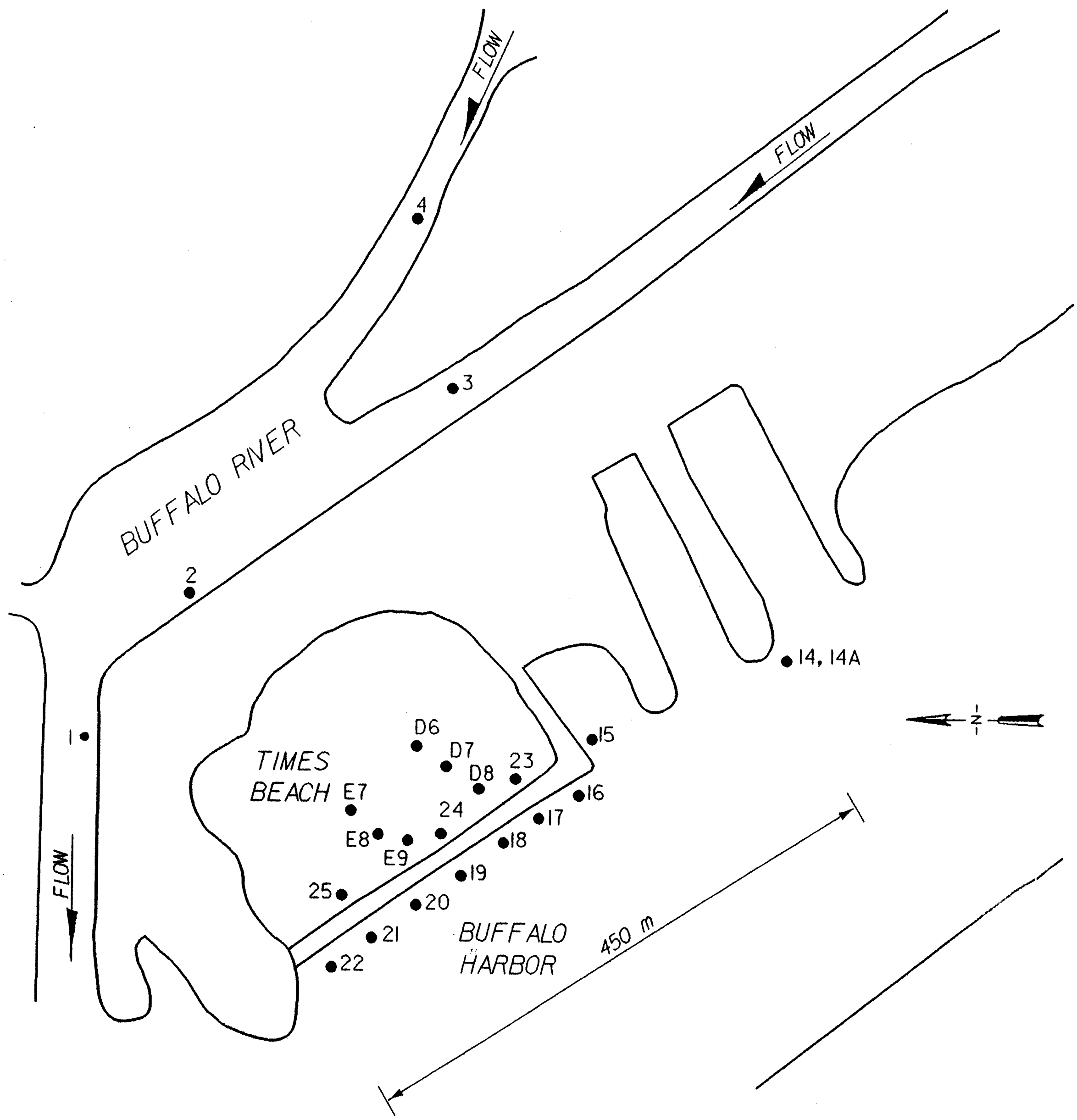

Figure 5. Biomonitoring locations in the immediate vicinity of the Times Beach CDF (see Table 3)

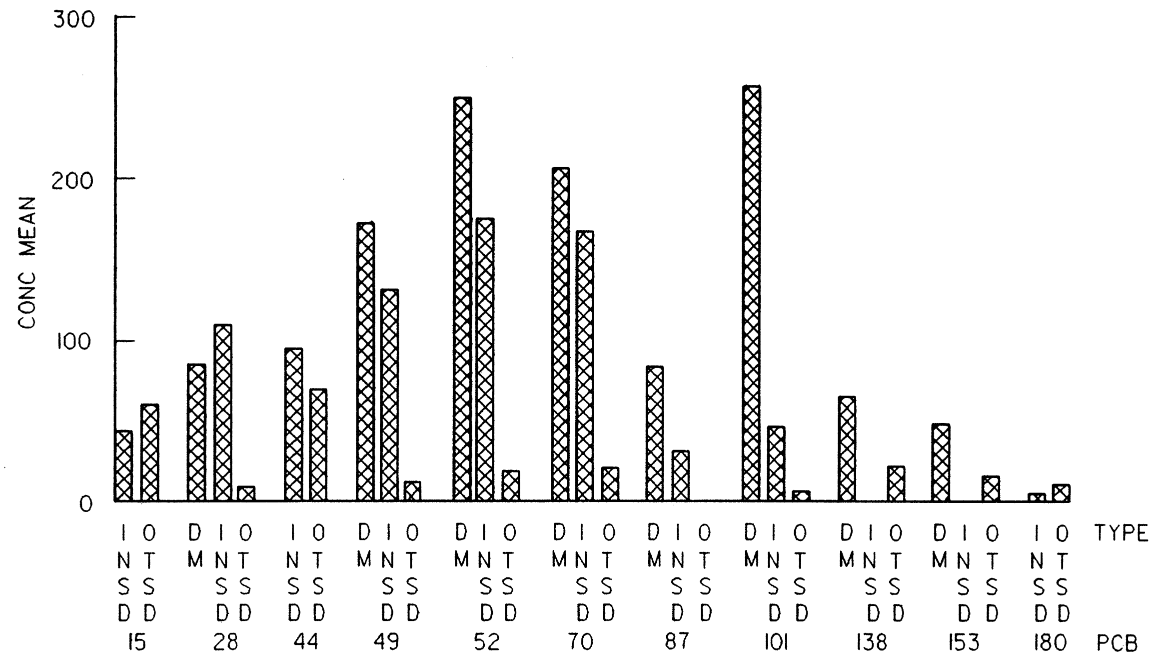

Figure 6. Mean concentrations of PCB congeners (IUPAC) in biomonitoring mussel tissues from inside (INSD) and outside (OTSD) the Times Beach CDF dike and in dredged material within the CDF (DM). Concentrations are in micrograms per kilogram ash-free dry weight for tissues and micrograms per kilogram dry weight for dredged material